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Dr. Valerii M Cherepanov

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14. ABSTRACT

This report results from a contract tasking Russian Research Center Kurchatov Institute as follows: This experiment is directed toward the observation of a perturbation of the Mossbauer spectrum of a nuclear isomer through the influence of an intense optical-frequency field. It could lead to laser control of nuclear transitions, and would open up a new area of research. Firstly, laser-Mossbauer spectroscopy would offer a vast range of applications starting from very fundamental, such as the first direct measurement of nuclear radii in excited atoms. Secondly, it will be very useful for practical applications, such as characterization of optical materials heretofore resistant to Mossbauer probing, or the studies of various dopants and their environment in different crystalline hosts.

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Final
Project Technical Report
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Double Optical-Gamma Resonance

(From 1 October 2003 to 31 December 2004 for 15 months)

Valeriy Mikhailovich Cherepanov
(Project Manager)

Russian Resarch Center «Kurchatov Institute»

February 2005

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The major goal of this project was to make a proof of experiment demonstrating the changes in the Mossbauer spectra of ^{151}Eu nuclei under the action of the resonant laser pumping.
A complex laser-Mossbauer experimental set-up has been developed for studies of possibilities to observe the double optical-gamma resonance in the crystals activated by Mossbauer ions with a narrow optical absorption line. The set-up included a tandem “pump-laser – dye-laser” with about 1 W power of the continuous radiation in the range 550-620 nm, a Mossbauer spectrometer, a helium cryostat and needed attending equipment. At the given set-up technical characteristics the measured Mossbauer spectra of ^{151}Eu nuclei in the EuP_5O_{14} and $EuCl_3{\cdot}6H_2O$ crystals at room, liquid nitrogen and helium temperatures revealed only small (~3%) changes of the spectra line-width. Apparently, this result can be explained by the obstacle that the $^7F_0$ – $^5D_0$ optical transition line-width (~ 1 GHz) proved to be much larger than that of the laser (~10^{-1} GHz). The experiments on the searching for the double optical-gamma resonance will be continued on the crystals with narrower optical transitions and using a pulse laser.

Keywords: Experiment, Double resonance, Mossbauer spectroscopy, Laser, Pumping, Crystal, Temperature, Line-width.

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* 123182, Moscow, Kurchatov sq., 1, Russia
Phone +7(095)196-7786, Fax: +7(095)194-1994

E-mail: cherepanov@imp.kiae.ru

The work has been performed by
the following institutes.

1. Participated institutes:
    1.1 Leading institute:
Russian Research Center «Kurchatov Institute» (RRC KI)
123182, Moscow, Kurchatov sq. 1, Russian Federation
Phone +7-(095) 196-9645, Fax +7-(095) 192-8194
E-mail: ray@www.kiae.ru
    1.2 Subcontracted institute:
Lebedev Institute of Physics, Russian Academy of Sciences (LIPAS)
Phone +7-(095) 132-6346, Fax +7-(095) 938-2251
E-mail: sautkin@sci.lebedev.ru

II. Project Technical Report

2. Technical contents

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Introduction

(Brief description of the work plan: objective, expected results, technical approach)

So far, the possibility of Double Optical-Gamma Resonance (DOGR) was considered only theoretically. It was established that optical laser radiation can be used to manipulate nuclear transitions at gamma-ray energies and hence the Mossbauer absorption spectrum [1]. Therefore, it is particularly interesting to realize experimentally this specific double resonance technique, combining the advantages of the most precise tools in modern physics – laser and Mossbauer spectroscopy.

The project refers to the «Basic research» category and has the search character - to discover the double optical-gamma resonance effects theoretically predicted. Depending on the level of the power of pumping laser the following effects have been predicted:

1) changes in the population of the hyperfine sublevels and nuclei polarization,
2) changes in the isomer shift for a Mossbauer transition,
3) Rabi-splitting in Mossbauer spectra.

The objective of this project is to make a proof of principle DOGR experiment demonstrating the changes in the Mossbauer spectra of ^{151}Eu nuclei (as a reserve variant - ^{57}Fe nuclei) under the action of the resonant laser pumping.

In this project we suggested:

1) To carry out the DOGR experiment on ^{151}Eu in self-activated europium pentaphosphate (EPP) EuP_5O_{14} and europium trichloride (ETC) $EuCl_3{\cdot}6H_2O$ crystals in which the Eu^{3+} ion is not an impurity but a host element. It provides much greater Mossbauer absorption as compared to the absorption of strongly diluted paramagnetic impurity Mossbauer atoms in diamagnetic host crystals. Moreover, a very narrow $^5D_0$ - $^7F_0$ electronic transition of Eu^{3+} ion is conveniently located at 578 nm and can be effectively driven by a dye-laser [2, 3]. Therefore, Eu^{3+} has the most promising characteristics both for optical and Mossbauer parts of the experiment. Besides, ^{151}Eu is one of the most widely used (the third after ^{57}Fe and ^{119}Sn) Mossbauer elements, whose changes in the electronic state under laser pumping might be observed in the Mossbauer spectrum.

2) In the case of absence of the appreciable DOGR-effect on ^{151}Eu – to continue the experiment with ^{57}Fe impurity in ZnS crystals. It is well known that ^{57}Fe is the most suitable Mossbauer isotope. In addition, the Fe^{2+}-doped compounds (such as very pure semiconductors with zinc blende structure) are widely used in optical studies. Moreover, there is a suitable sharp zero-phonon optical line near 500-600 nm corresponding to the ^5E - ^3H electronic transition [4].

The proof-of-principal experiment on laser control of nuclear transition would open up a new area of research. Firstly, it would be laser-Mossbauer spectroscopy, with its vast range of applications starting from fundamental, such as the first direct measurement of nuclear radii in excited atoms. Secondly, it would be very useful for practical applications, such as characterization of optical materials heretofore resistant to Mossbauer probing, or the studies of various dopants and their environment in different crystalline hosts. Not the least, a potential outgrowth of this field could be a realization of gamma-ray laser (a coherent source of radiation with extremely short wave length, below 1 angstrom).

Conceptually the DOGR experiment is very simple. First, one needs to record conventional Mossbauer spectra of a sample in the absence of the resonant laser field, and then to record the modified spectra in the presence of laser radiation. The difference between these spectra will reveal the looked for DOGR-effect, which should be a subject of a theoretical analysis.

To provide the needed conditions for the DOGR experiment we solved the following tasks:
1) To build up a complex laser-Mossbauer experimental set-up;
2) To modify a cryostat in order to pass both laser and gamma beams, and to prepare specimens;
3) To measure the DOGR spectra of ^{151}Eu nuclei in EuP_5O_{14} and/or $EuCl_3{\cdot}6H_2O$ crystals at room, nitrogen and helium temperatures and publish the results;
4) In the case of absence of the appreciable DOGR-effect on ^{151}Eu – to growth ZnS: $^{57}Fe^{2+}$ crystals to measure the DOGR spectra of ^{57}Fe impurity at helium temperature and publish the results.

Methods

1) Creation of the experimental set-up

a) A special basement has been made for the SP-171-18 argon pump-laser and the SP-380D dye-laser. A new electrical wiring of the three-phase alternating voltage 380 V with the current up to 60 A was additionally fed to supply the lasers. A water system for the laser cooling was installed in the apartment. A test switching on the lasers has revealed that the pump-laser power supply was out of order. The spoilt elements (mainly powerful diodes of the output stage) of the unit electric scheme were found and replaced with the new ones. Then the pump laser mirror system was aligned and normal operation of the lasers was achieved. A tuning of the tandem «argon laser SP-171-18 – dye-laser SP-380D» to the EPP crystal main absorption line 578.2 nm was fulfilled. The output power of the laser was about ~1 W, and the power absorbed by the EPP-sample was estimated ~10^{-2} W.
An additional basement with a part of the Mossbauer spectrometer, including the driver with the radioactive source ^{151}Sm of about 30 mCi activity and the scintillation counter, was placed near the set-up with lasers. Axes of the optical and gamma beams were aligned at the right angle one to another.

b) An additional experimental set-up - neodymium laser with the second harmonic pumping (λ=532 nm) was assembled to measure the optical absorption spectra in the range of 560-620 nm. The set-up included a Rodamine-6G dye-laser with an amplifier, a nanosecond laser photometer, a lambda-meter, an integrator, a programmable saw-generator, a digital voltmeter, a digital oscillograph, a system of beam-splitters and ordinary mirrors, delay lines and phase detectors. The Nd-laser had the pulse repetition rate of 20 Hz and the pulse energy up to ~20 mJ. About 8 mJ of the pulse energy was focused by a cylindrical lens into the cell of the dye-laser (transverse pumping) and the rest ~12 mJ with ~2 ns delay was focused into the amplifier cell (longitudinal pumping). A duration of the pumping pulse was ~10 ns. The dye-laser included a cell, a cylindrical lens, an output mirror, a diffraction lattice (2400 lines/mm), an ordinary mirror and a prismatic telescope. Dye-laser tuning to the wavelength of interest was realized by the ordinary mirror turn with a help of a micrometer screw. The fine-tuning near the proper value of λ was made by moving the mirror with a

piezoelectric transducer, controlled by a ramp pulse (0 - 500) V from the programmable generator. The laser wavelength was measured with the λ-meter. A time-averaged generation line width was found to be ~2 GHz and consisted of 3 - 4 modes. The laser beam diameter was ~6 mm, and the pulse energy measured by the laser nanosecond photometer was about 160 mkJ.

c) The previous sample holder in the available helium cryostat was cut off. The new sample holder was made and installed to allow for rotation of the sample plane at the angle 45^o to the gamma-quanta beam. The two additional windows in the lower cryostat cover were made to pass the laser beam. The direction of laser beam was also chosen to be at the angle 45^o to the surface of the sample. The windows were made from the quartz glass with thickness 1 mm and diameter 15 mm.
After the first attempt to carry out the DOGR measurements at low temperatures the cryostat was again modified in order to minimize the distance between the Mossbauer source and counter and to raise the gamma-quanta counting rate. For this, all the elements of the bottom part (the specimen holder, the thermal screen and the external shell with the optical and gamma windows) were made about two times smaller. As a result, the gamma-quanta counting rate was raised about three times.

2) Preparing of specimens

a) Europium pentaphosphate.
The largest and most transparent EPP crystals from the available batch of crystals were selected. They were cut to the thickness of about 1 mm and were polished down to the thickness of 0.5 mm. A mosaic target of about 9x6 mm^2 in cross section from ten such crystals was mounted on a transparent plastic disk covered by a viscous liquid for measurements of optical absorption- and Mossbauer spectra. The rest small crystals were milled into a powder in order to prepare a Mossbauer absorber of larger area.

b) Europium trichloride hexahydrate.
A big ETC single crystal with sizes 10x7x2 mm^3 was grown from the saturated water solution in the Institute of Applied Physics RAS (N.Novgorod, Russia) and was kindly offered by prof.O.Kocharovskaya. Due to its high hygroscopicity it was put on an Al-foil with a small hole for passing the fluorescence radiation and hermetically packed in a transparent plastic cell.

c) Zinc blende.
A new technique of growing of ZnS crystals doped by iron-57 isotope with concentration up to 0.2% has been developed in LIPAS since the production of such crystals of rather high optical quality has not been realized in RF earlier. In the process of the zinc blende crystal growth iron should be in the form of the two-valence iron compound FeS. For this, we adjusted the method of the metal iron restoration in the hydrogen flow at temperature 900 C at first. A small part of a substance produced was crushed into powder to prepare the Mossbauer absorber. The parameters of the Mossbauer spectrum of iron-57 nuclei (the values of isomer shift, quadrupole splitting and effective magnetic field) measured at room temperature showed full coincidence with those known from literature for iron sulfide FeS.
We have succeeded to grow a big ~15x10x10 mm^3 crystal which was cut into five parts of ~1 mm thickness. From each crystal, small pieces with mass of about 40 mg were cut to prepare polycrystalline Mossbauer absorbers. The measurements of Mossbauer absorption spectra on ^{57}Fe nuclei showed that iron impurities were distributed in the initial crystal volume inhomogeneously. The iron concentration in the top part of the crystal seemed to be so large that even magnetic hyperfine structure from six broadened lines was found in the spectrum. At the same time, for the bottom crystal part the Mossbauer spectrum consisted from a single line, corresponding Fe^{2+}, the parameters of which were close to the known literature data for the doped ZnS crystals. However, the iron concentration appeared to be so small that the Mossbauer absorption was about 0.3%, which was not enough to carry out the main measurements. Studies of the rest crystals and the

following adjusting of the crystal growth procedure also showed that the iron concentration appeared to be an order smaller than it needs for the DOGR experiment because the Mossbauer absorption seemed to be only about 0.6%, which was again not enough to carry out the main measurements.
At the present, we are continuing the attempts to grow the suitable crystal of the iron doped zinc blende.

Optical measurements

a) For the optical quality estimation of small (about 3x2x1 mm^3) EPP crystals, which have monoclinic symmetry, their optical absorption spectra in the range 560-620 nm have been measured. A spectrophotometer, made on the base of the monochromator MDR-23 with the resolution of 0.02 nm was used. For the 578.2 nm line of interest (the transition from the Eu^{3+} ion ground state $^7F_0$ to the first excited state $^5D_0$) the absorption value about 4% was obtained for the crystal thickness of 1mm. The absorption is small since this type of transitions $<A_1 - A_{1>}$ is forbidden for the cubic symmetry of a crystal and becomes weakly allowed as the symmetry is violated. On the other hand, such a «semi-forbidden» character of the optical transition provides a comparatively long lifetime of the Eu^{3+} excited state and, correspondingly, a small line-width of the transition which is important for the double resonance observation.

b) The emission optical spectra were measured by using excitation with 60 keV X-Ray radiation from the X-Ray tungsten tube. The fluorescence spectra (Fig. 1) revealed the well defined line at 578.2 nm (weak), the characteristic triplet $^5D_0$ - $^7F_1$ with wavelengths 587.3, 592.0 and 594.7 nm (more intensive), and the most intensive 611.5 nm line from the $^5D_0$ - $^7F_2$ quintet.

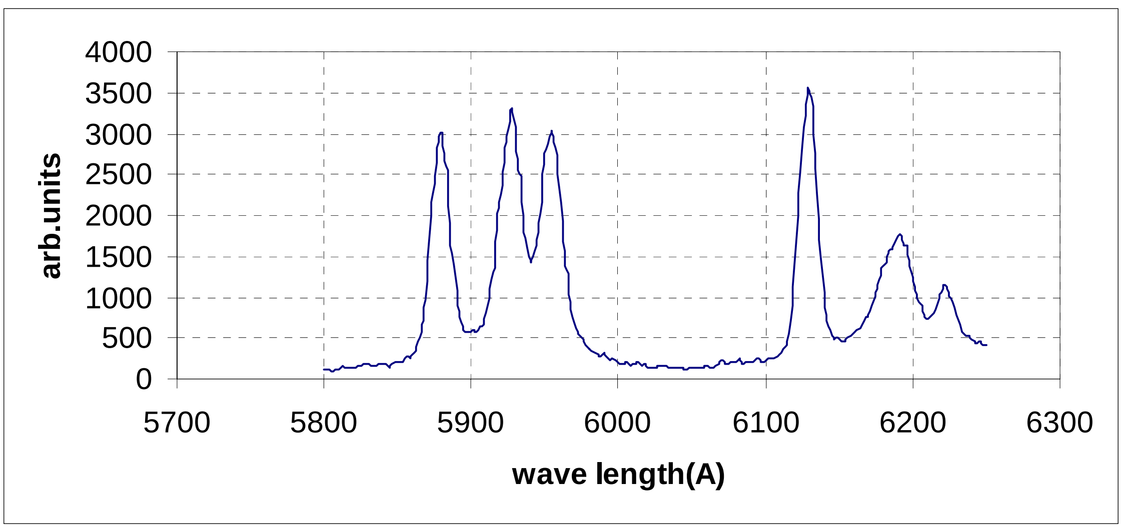

Fig. 1. Fluorescence optical spectrum of EuP_5O_{14} single crystal at room temperature under the 60 keV X-Ray excitation measured with the monochromator MDR-23.

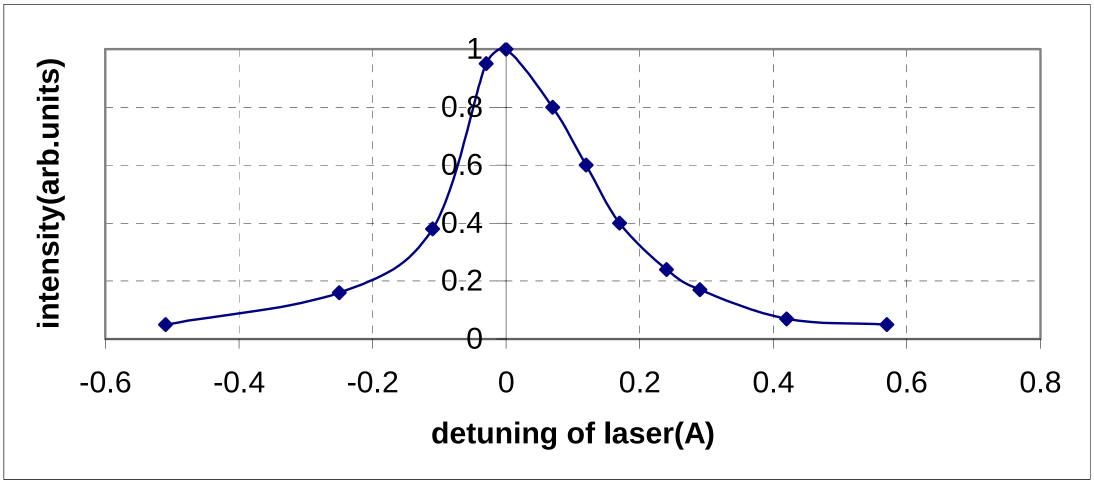

Fig. 2. Dependence of the fluorescence signal at the wave-length 611.5 nm on the dye-laser detuning. It corresponds to the absorption line-shape of 578.2 nm at room temperature in the air. Solid line is a guide for eyes.

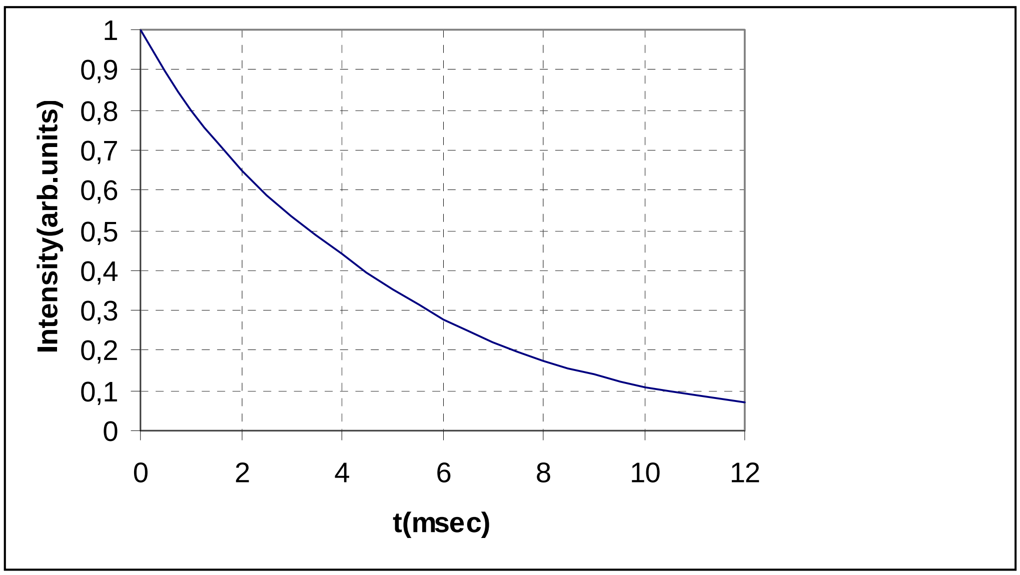

Fig. 3. The decay curve of the $^5D_0$ excited state of Eu^{3+} ions in the EuP_5O_{14} single crystal at T=295K after excitation by the pulse laser on the 578.2 nm with pulse energy 20 mJ, repetition frequency 20 Hz and pulse duration 10 ns. The life-time of the level estimated as t_0=4.7(3) ms.

For the measurements of the fluorescence spectra and the excited-state lifetime, we used the pulsed dye-laser pumped by the second harmonic (λ=532 nm) of a Nd-laser. The pulse repetition rate, duration, and energy were 20 Hz, ~10 ns, and about 10 mJ, respectively. The dye-laser was tuned to the main transition at 578.2 nm. The spectra were displayed with a monochromator followed by the PMT-79 photomultiplier. For the exciting spectra measurements, monochromator was tuned to a fluorescence line (in our case 611.5 nm), and the dye-laser wavelength was scanned near 578.2 nm. At room temperature, the exciting line-width was found to be 0.023 nm (Fig. 2), and the measured value of the lifetime of the $^5D_0$ level was estimated as t_0=4.7(3) ms (Fig. 3). The dependence of the fluorescence signal (at 611.5 nm) on the surface density of the pulse energy, P, of the dye-laser tuned to the exciting line maximum was also measured.

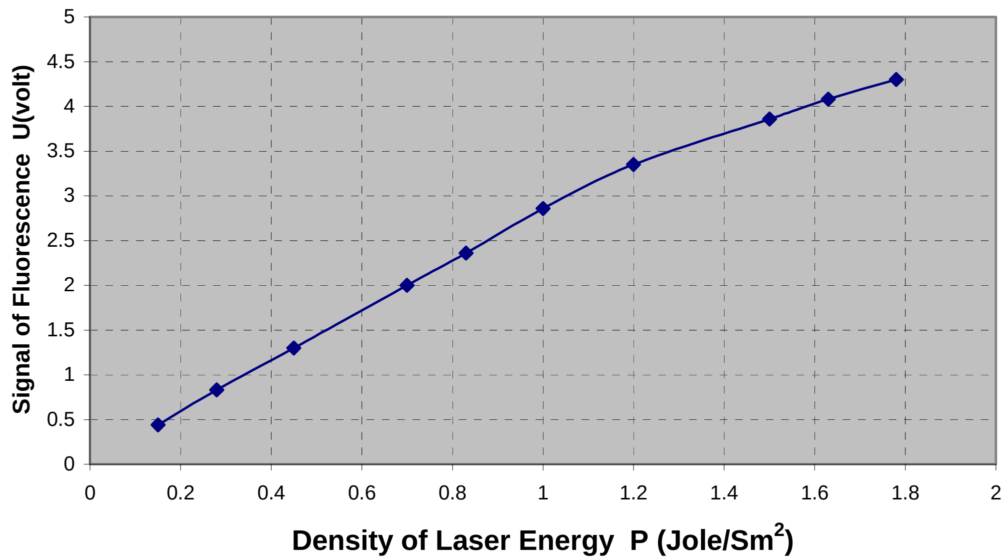

Fig. 4. Dependence of the fluorescence signal on the surface density of laser energy, P, at room temperature. The saturation starts at about P~1.2 J/cm^2.

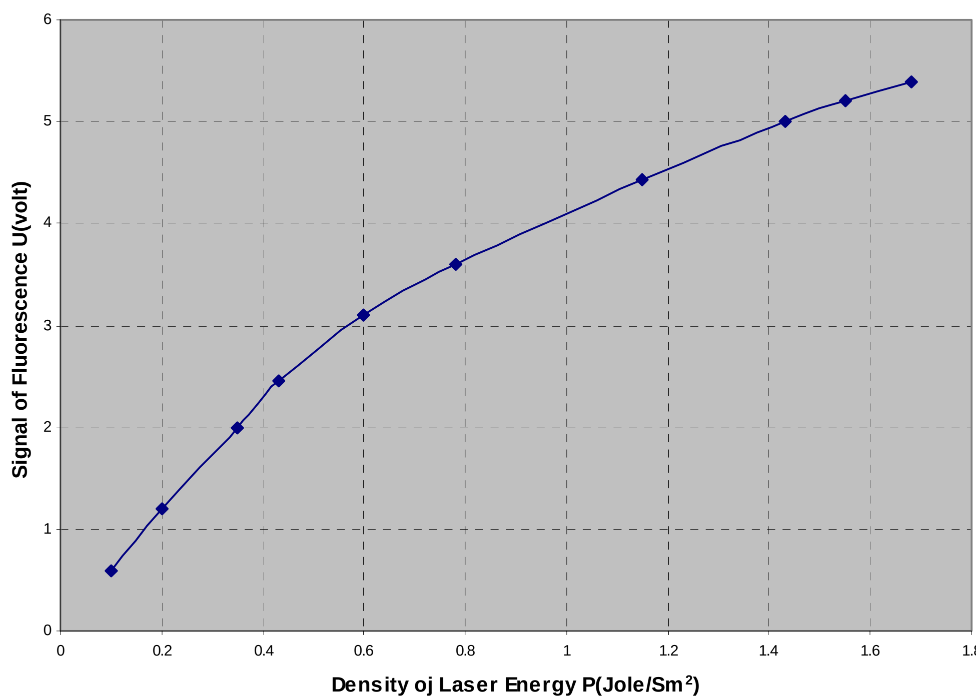

Fig. 5. Dependence of the fluorescence signal on the surface density of laser energy P at T=80K. The saturation starts at about P~0.4 J/cm^2.

At room temperature the fluorescence signal increased linearly up to P≈1.2 J/cm^2, then the saturation started to appear (Fig. 4). At T=80 K the saturation started at P≈0.4 J/cm^2 due to the narrowing of the absorption line (its length increased to 578.4 nm (578.6 nm in vacuum)) of about three times in comparison with the room temperature value (Fig. 5).
The ETC single crystal optical spectra measurements showed the similar results with the following values of the wave-length: the main transition 580.2 nm and the fluorescence line 613.3 nm.

<u>Mossbauer measurements</u>

a) The Mossbauer spectra of europium-151 nuclei of the prepared polycrystalline EPP and ETC samples were measured at room and liquid nitrogen temperatures. Its showed that all europium ions are three-valence and the two-valence species are not detected in the limits of approached statistical accuracy of 0.1%. This observation confirms a chemical homogeneity of the materials. Moreover, the spectrum shape represents itself as a slightly asymmetric broadened single line, indicating the presence of a small quadrupole splitting whose magnitude is much smaller than the doubled Mossbauer natural line-width $2G_n$=1.31 mm/s. This is a consequence of low monoclinic symmetry of oxygen ions coordination of the nearest europium atom surroundings. Note, that as far as we know, the europium pentaphosphate Mossbauer spectrum is measured at the first time.

The measurements of the EPP and ETC single crystals were performed also at T=295 K (Figs. 6, 7) and T=80 K. The latter spectra, just as the former ones, also represented an asymmetrical single line with the unresolved quadrupole splitting. Note, that because of the ^{151}Eu nucleus has large spins (I_g=5/2 and I_e=7/2 for the ground end excited states, respectively) the Mossbauer spectrum is complex and consists of 8 lines allowed by the selections rules (12 lines in the case of the symmetry lower than axial). However, usually the quadrupole interaction constant is small in comparison with the line-width and the simplified single line representation can be used for just search for any changes in spectra under laser pumping.

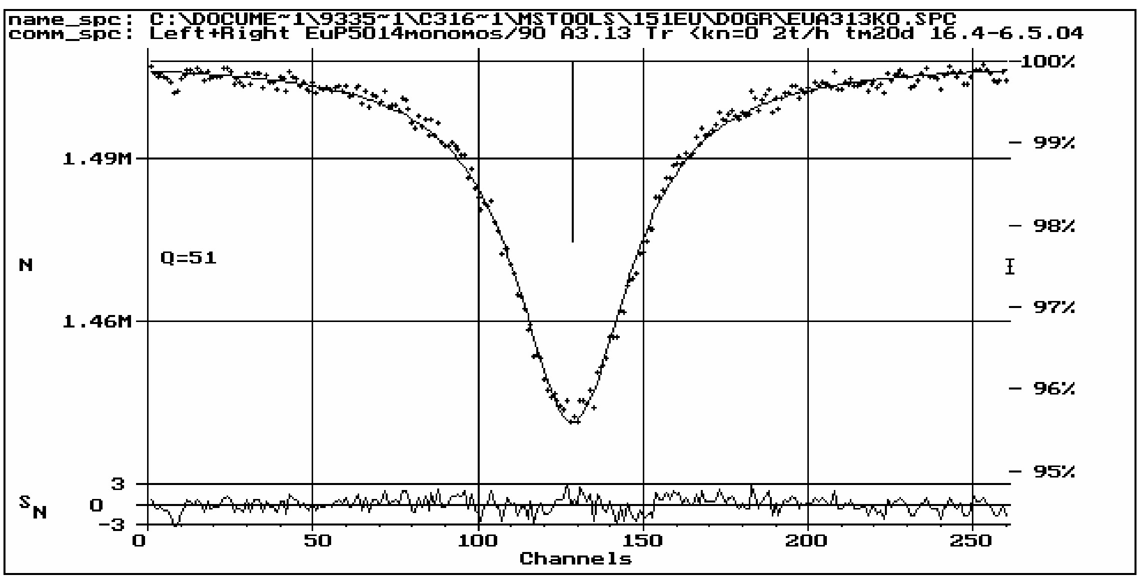

Fig. 6. Mossbauer spectrum of the europium pentaphosphate (EPP) EuP_5O_{14} single crystal mosaic at T=295K. Solid line is the result of the fit in the lorentzian approximation in the simplified single line representation of the spectrum shape. The zigzag line below is the discrepancy between the experimental and calculated spectra.

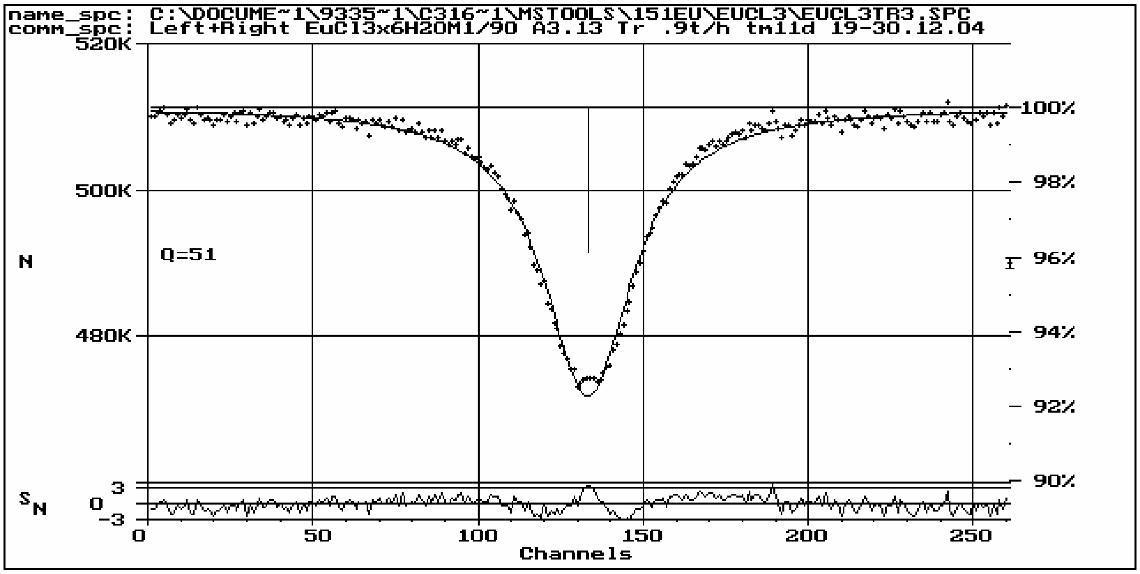

Fig. 7. Mossbauer spectrum of the europium trichloidee (ETC) $EuCl_3{\cdot}6H_2O$ single crystal at T=295K. Solid line is the result of the fit in the lorentzian approximation in the simplified single line representation of the spectrum shape. The zigzag line below is the discrepancy between the experimental and calculated spectra.

b) First, the simulation experiment on the DOGR was performed at room temperature on the EPP crystal. The sample was formed as a mosaic from ten EPP crystals on a transparent plastic substrate and mounted at the angle 45^o to the both beams. The distance between the source and the counter proved to be about 10 cm, and the counting rate of the Mossbauer gamma quanta with the energy 21.6 keV was about 10^3 pulses/hour.channel. At this time a tuning of the tandem «argon laser SP-171-18 – dye-laser SP-380D» to the EPP crystal main absorption line (578.4 nm in vacuum) was fulfilled. The tuning to this line was monitored by measuring of the optical fluorescence maximum on the wavelength 611.5 nm and was produced manually with an accuracy of about 10% of the maximal signal.

The Mossbauer spectra, measured with and without laser resonant pumping on the wavelength 578.2 nm at the small intensity ~0.1 W/cm^2, were recorded alternately into different memory parts of the Mossbauer spectrometer. A pure measuring time for each spectrum was about one week. In both cases, the Mossbauer spectra represented the slightly asymmetrical broadened line evidencing the presence of a small quadrupole splitting, just as in the case of the polycrystalline sample. At the same time, the Mossbauer spectra obtained with and without pumping coincided with each other in the limits of the statistical accuracy.

Results

1) Measurements of the DOGR spectra in EuP_5O_{14} crystal.

a) Measurements of Mossbauer spectra with and without laser excitation at room temperature.

After the model experiment on DOGR the direct measurements of the sought effect at room temperature have been started with the mosaic EPP specimen mounted in the copper holder of the cryostat «cold finger» on a transparent plastic substrate in vacuum. In this case the distance between the source and the counter proved to be about 20 cm, and the counting rate of Mossbauer gamma quanta reduced down to about $3{\cdot}10^2$ pulses/hour.channel. The intensity of the dye-laser in the single mode regime was about ~1 W/cm^2 (at the optical and gamma beam diameter ≈6 mm), and the intensity absorbed by the EPP-sample was estimated to be about ~10^{-2} W/cm^2 from the

optical absorption spectra measurements. In the DOGR measurements, the Mossbauer spectra were recorded with and without laser resonant pumping into different memory parts of the Mossbauer spectrometer alternately at 4 hours in each regime.

Then the DOGR measurements at room temperature were continued in the multimode regime with the line-width of about 150 MHz of the dye-laser until the argon-laser failed. A pure measuring time for each spectrum again was about one week at the counting rate of Mossbauer gamma quanta about $3\cdot10^2$ pulses/hour.channel. However, as just as in the model experiment, the Mossbauer spectra obtained with and without pumping also coincided with each other in the limits of the statistical accuracy. The Mossbauer line parameters (without taking into account the hyperfine quadrupolar splitting, for the resolution of which a statistical quality of the spectra was still insufficient) appeared to be the following (in the mm/s):

the isomer shift IS_1=-0.11(1), the line-width G_1=3.43(6), and
the isomer shift IS_2=-0.11(1), the line-width G_2=3.38(6)
for the spectra with (1) and without (2) laser excitation, respectively.

A conclusion was drawn that the DOGR effect for ^{151}Eu nuclei in the EPP crystals is not observable at room temperature at the available dye-laser power although the line-width value G_1 in the former case is about 1.5% larger than in the latter one.

b) Mossbauer spectra taken with and without laser excitation at liquid nitrogen temperatures.

During a month, because of the argon-laser has been failed to continue the measurements, we had found, mounted and tuned the substitution for it: an analogous argon-laser of the SP-171-19 model. The output power and other characteristics of the laser were practically matched to those of the previous laser of the SP-171-18 model.

At first, for the low temperature DOGR measurements the mosaic specimen EPP was mounted in the copper holder of the cryostat «cold finger» on a transparent plastic substrate. The Mossbauer spectra of DOGR were measured at T≈150K (due to low thermal conductivity of the substrate) in the single-mode regime of the dye-laser at the output power of about ~1 W on the 578.3 nm. Again, the pure measuring time for each spectrum was about one week. The Mossbauer line parameters appeared to be the following (in the mm/s):

the isomer shift IS_1=-0.08(2), the line-width G_1=3.75(9), and
the isomer shift IS_2=-0.08(2), the line-width G_2=3.61(9)
for the spectra with (1) and without (2) laser excitation, correspondingly.

After the further modification of the cryostat and attaching the mosaic EPP specimen to the Al-foil substrate (with a small ~1 mm hole for passing of the fluorescent radiation on 611.5 nm) the specimen temperature was reduced to T=80K. Additional measurements of the EPP optical absorption spectra showed that at the specimen temperature T=80K the main absorption line length has been raised to 578.4 nm, and its line-width decreased about three times in comparison with the room temperature value. Therefore, the optical absorption cross section has been increased at least as many times as the line-width has decreased. In this case, the DOGR measurements in the multimode regime showed the following Mossbauer line parameters (in the mm/s):

the isomer shift IS_1=0.02(2), the line-width G_1=4.16(8), and
the isomer shift IS_2=0.02(2), the line-width G_2=4.03(8)
for the spectra with (1) and without (2) laser excitation, correspondingly.

A conclusion was drawn that 3% broadening of the Mossbauer line-width has been observed for ^{151}Eu nuclei in the EPP crystals at T=80K and about threefold increase of the dye-laser power, which could be evidence of the sought Rabi-splitting presence. However, the statistical error in this case is too high in order to ascertain the DOGR-effect observation confidently.

c) Measurements of Mossbauer spectra with and without laser excitation at liquid helium temperature.

The change of the argon-laser resulted in a greater than ~1 month delay, therefore the DOGR measurements in the multimode regime at T=5K were performed only at the quarter-4. The results of all the temperature DOGR measurements on the EPP specimen are collected in the Table 1 below:

Table 1. Parameters of the ^{151}Eu Mossbauer spectrum of the EuP_5O_{14} single crystals in the single Lorentz line approach obtained in the DOGR measurements at different temperatures. IS – the isomer shift (mm/s), G – the line width (mm/s), $\Delta G=(G_1 - G_2)$, (1) and (2) – are taken with and without laser excitation, respectively.

T, K	IS_1	IS_2	G_1	G_2	$\Delta G/G$, %
295	-0.11(1)	-0.11(1)	3.43(6)	3.38(6)	1.5
150	-0.08(2)	-0.08(2)	3.75(9)	3.61(9)	3.9
80	0.02(2)	0.02(2)	4.16(8)	4.03(8)	3.2
5	0.09(3)	0.01(3)	4.18(9)	4.07(9)	2.7

2) Measurements of the DOGR spectra in $EuCl_3 \cdot 6H_2O$ crystal.

All measurements of the Mossbauer spectra with and without laser excitation at room and liquid nitrogen temperatures were performed at the quarter-5 under the same conditions as in the case of the EPP crystal (the room temperature measurements were carried out without the cryostat). The results of all the temperature DOGR measurements in the multimode regime on the ETC specimen are collected in the Table 2 below. Note, that the ETC results slightly differ from that of the EPP.

So, at room temperature, the line-width values G_1 and G_2 coincide between one another but substantially smaller than that of the EPP, evidently due to the smaller ETC quadrupole splitting magnitude. With temperature decreasing to T=80K, the line-width values G increased in the same extent as in the EPP case, and the G_1 value is noticeable larger than that of the G_2. Meanwhile, the IS_1 value is slightly reduced in comparison with that of the IS_2. This could be a consequence of the small sample warming during the laser pumping because of too low heat transfer in the cryostat.

Table 2. Parameters of the ^{151}Eu Mossbauer spectrum of the $EuCl_3 \cdot 6H_2O$ single crystal in the single line approach obtained in the DOGR measurements at different temperatures. All the notations are the same as in the Table 1.

T, K	IS_1	IS_2	G_1	G_2	$\Delta G/G$, %
295	0.27(1)	0.26(1)	2.62(2)	2.61(2)	0
80	0.31(1)	0.33(1)	3.20(9)	3.00(9)	6.2

Conclusion

On the results of all the DOGR measurements the following conclusions can be made:

a) The line width G_2 (without excitation) increases with temperature decreasing for both specimens. This can be explained by the quadrupole splitting growth, which is typical for rare earth Mossbauer nuclei;

b) Up to T=80K the isomer shift values IS_1 and IS_2 practically coincide within the accuracy limits that confirms the absence of the essential warming of the specimen during the laser pumping;

c) At T=5K the isomer shift value of the EPP at the laser pumping, IS_1, is noticeable larger than that without pumping (IS_2) and moves to the increased europium ion valence Eu^{4+}. As the sample warming must lead to the isomer shift decreasing this fact could be the evidence of noticeable population of the excited $^5D_0$ orbital singlet and $^7F_1$, $^7F_2$ etc. multiplets of the Eu^{3+} ion ground state during the laser resonant pumping.

d) At low temperatures a systematic increase of the Mossbauer line-width G_1 growth under pumping (in average about 4%) is observed. It could also evidence a small quadrupole splitting growth due to the same origin or indicate the presence of a sought small Rabi-splitting predicted of the DOGR theory. However, the statistical error in this case is too much high in order to ascertain the DOGR-effect observation confidently.

e) Apparently, this result can be explained by the obstacle that in spite of the long life-time (t_0~5 ms) the $^7F_0$ – $^5D_0$ optical transition line-width (~ 1 GHz) proved to be much larger than that of the laser (~10^{-1} GHz). The experiments on the searching for the double optical-gamma resonance will be continued on the crystals with narrower optical transitions and using a pulse dye-laser. Therefore, the following attempts to observe DOGR at low temperatures in the multimode and pulse regimes will be undertaken on another crystals: possible candidate – europium hydroxide $Eu(OH)_3$. The latter has exceedingly narrow (~0.17 GHz) the $^7F_0$ – $^5D_0$ transition.

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List of published reports and presentations at conferences with abstracts

1. V.Cherepanov “Mossbauer spectroscopy of electron-nuclear transitions”, Mini-Symposium “Gamma-Ray Optics and Quantum Nucleonics” of the II International Conference “Frontiers of Nonlinear Physics” (N.Novgorod – St.-Petersburg, Russia, July 5-12, 2004, Abstracts, p.114) sponsored by EOARD.

Abstract of the report see below

MOSSBAUER SPECTROSCOPY OF ELECTRON-NUCLEAR TRANSITIONS

V.M.Cherepanov

Russian Research Center Kurchatov Institute, 123182 Moscow, Russia

Two types of mixed electron-nuclear transitions will be discussed: (1) spontaneous and (2) induced. For paramagnetic atoms diluted in a diamagnetic host crystal the hamiltonian can be written as

$$\boldsymbol{H}=\boldsymbol{H}_{GT}+\boldsymbol{H}_{CF}+\boldsymbol{H}_{HF}+\boldsymbol{H}_{Z}+\boldsymbol{H}_{Q}+\boldsymbol{H}_{R}, \qquad (*)$$

the terms are respectively: the Mossbauer gamma-transition, the crystal field splitting, the hyperfine structure (HFS), the Zeeman interaction (both for the electron and nuclear spins), the quadrupole interaction and the lattice (relaxation) term. The observability of the electron-nuclear transitions depends on the rates at which energy can be transferred between the various energy levels represented by Eq.(*). For the most convenient Mossbauer nucleus ^{57}Fe the lifetime of the excited nuclear level is $T_n \approx 10^{-7}$ s, while that of the excited optical level is typically in the range $10^{-8} - 10^{-2}$ s. These are of the order of the relaxation times for NMR and ESR transitions.

Fe^{3+}-ion has a $3d^5$, 6S electronic state, which is favourable for ESR studies, and its nuclear spins ($I_g=1/2$, $I_e=3/2$) are favorable for NMR. ESR, NMR and Mossbauer Spectroscopy (MS) enable electronic or nuclear spin subsystems to be studied separately. ENDOR and NMR-Mossbauer double resonance (DGMR) allow one to study interactions of the electronic and nuclear subsystems in the ground and excited nuclear states, respectively. So far, aluminum nitrate $Al(NO_3)_3 \cdot 9H_2O{:}Fe^{3+}$ is the unique substance where the transitions both (1) and (2) types were observed experimentally.

(1) The $^6S_{5/2}$ ground state of Fe^{3+} ion in the crystal field is split into three Kramers doublets, one of which has an almost isotropic and two another ones have high anisotropic electronic g- and hyperfine A-tensors. The total Mossbauer spectrum then consists of three superposed subspectra corresponding to each doublet. When the direction of an external magnetic field H_o~100 G is close to the plane where lie the smallest g-tensor components of one of the anisotropic doublet intensive extra-lines (Z-lines), lying far beyond the range of Doppler velocities typical for Fe^{3+}, were found [1]. The physical origin of the lines is as follows. In a weak magnetic field (respective to the crystal field Stark splitting about 1K in aluminum nitrate) the electronic Zeeman energy is comparable to the energy of the magnetic hyperfine interaction. In this case both the ground and the excited nuclear states have a mixed electron-nuclear structure. If the nuclear transition alters the electronic state as well as the nuclear one, the Zeeman interaction may supply some of the energy needed for the transition.

(2) The possibility of double (a) MS+NMR, (b) MS+ESR or (c) MS+Laser resonances performing has been discussed since 60-s.

(a) Most of experiments on the double gamma-magnetic resonance (DGMR=MS+NMR) were realized on ferromagnets [2]. As for paramagnets, the first experiment on DGMR was performed on aluminum nitrate doped with ^{57}Fe [3], where a noticeable broadening of lines for isotropic Kramers doublet subspectrum was induced by the intensive rf magnetic field H_{rf}~10G at the resonant frequency for the excited nuclear state.

(b) In our experiment [4] on the double gamma-electronic resonance (DGER=MS+ESR) the Mossbauer spectra of 0.5% $^{57}Fe^{3+}$ impurity in aluminum nitrate single crystal measured with and without ESR-excitation (T=70K, P=50mW, F=9.41GHz, H_o=96G, H_{rf}~0.7G - the case of the mixed electron-nuclear transitions - Z-lines) revealed two effects. The first one is a marked broadening (by~1.3 times) of one of the Z-lines. The second one is a small broadening of the isotropic Kramers doublet subspectrum.

(c) Possibilities of the double gamma-optic resonance (DGOR=MS+Laser) was analyzed in [5].

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Project manager